

Phase-sensitive-measurement determination of odd-parity, spin-triplet superconductivity in Sr_2RuO_4

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Phase-sensitive-measurement determination of odd-parity, spin-triplet superconductivity in Sr_2RuO_4

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Abstract. In this paper, I present a brief summary of the physical properties of Sr_2RuO_4 and also review our work on the Josephson effect and phase-sensitive measurements of Sr_2RuO_4 . Our results provide strong support to the prediction that this material is an odd-parity, spin-triplet superconductor. I also discuss the eutectic phase of $\text{Ru-Sr}_2\text{RuO}_4$ and comment on several unresolved issues regarding Sr_2RuO_4 .

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1. Introduction

Superconductivity occurs because of the formation of Cooper pairs. The Fermi statistics of electrons demands that the wave function of a Cooper pairs is asymmetric with respect to interchanging the individual coordinates (\mathbf{r}_1 and \mathbf{r}_2) and spins (s_1 and s_2) of the two electrons in the pair. For a superconductor with a translational invariance, the wave function of a Cooper pair can be written as a function of the relative coordinate, $\mathbf{r}(=\mathbf{r}_1 - \mathbf{r}_2)$, or the corresponding wave vector, \mathbf{k} , and the two spins. The interchange of the two electrons becomes inverting \mathbf{r} or \mathbf{k} plus the interchange of the spins. Therefore, under the inversion transformation, the Cooper pair wave function has to be either an even function (even-parity), with the total spin $S = 0$ (spin-singlet), or an odd function (odd-parity), with the total spin $S = 1$ (spin-triplet), to ensure that the total wave function is asymmetric with respect to particle interchange. As a result, superconductors can be divided into two categories—even-parity, spin-singlet superconductors or odd-parity, spin-triplet superconductors [1]. Superconductors falling into these categories can be further classified if additional symmetries exist. For example, in the presence of a rotational symmetry, the angular momentum quantum number, l , is a good quantum number. The spatial part of the wave function can be expressed by a spherical harmonic function. For even-parity, spin-singlet superconductors, l can be zero or an even number, leading to the familiar s-wave ($l = 0$) or d-wave ($l = 2$) superconductors. For odd-parity, spin-triplet superconductors, l can be 1 (p-wave) or 3 (f-wave) and, so on. For a crystalline superconductor, its pairing symmetry is classified according to the point group because the continuous rotational symmetry does not exist [1]. The superconducting order parameter is a scalar for spin-singlet and a vector for spin-triplet superconductors. A convenient form for the latter is the so-called d-vector, used in describing superfluid ^3He [2]. The magnitude of the d-vector represents the superconducting energy gap, while its direction is that perpendicular to the plane into which spins of the Cooper pairs are aligned.

Except for a few unusual classes of superconducting materials, most superconductors discovered to date, including all elemental superconductors, are s-wave superconductors. It is known that the pairing symmetry of the high- T_c superconductor is predominantly d-wave. Superfluid ^3He is the first experimentally established p-wave (charge neutral) superconductor [1]–[3]. The occurrence of superfluidity in ^3He is driven by the attractive interaction in the p-wave channel through spin fluctuations and their feedback effects [2]. Heavy fermion superconductors were the first serious candidate for electronic spin-triplet superconductivity [1, 4, 5]. The strong Coulomb repulsion in these heavy fermions appears to exclude significant attractive interaction in the s-wave channel and therefore an s-wave pairing. However, even though it is widely accepted that non-s-wave pairings prevail in heavy fermion superconductors, consensus on the exact pairing symmetry for any heavy fermion superconductor has proven to be difficult to establish [6]. After the discovery of superconductivity in Sr_2RuO_4 [7], which has an intrinsic superconducting transition temperature (T_c) of 1.5 K [8], and the subsequent prediction [9, 10] that the superconducting pairing symmetry in Sr_2RuO_4 is p-wave, it quickly became a leading candidate for establishing the long-sought spin-triplet superconductivity.

The p-wave pairing state in Sr_2RuO_4 was predicted based on the observation of some key properties of this material. Rice and Sigrist [9] suggested that the apparent $S = 1$ correlation in $\text{Sr}_2\text{Ir}_{1-x}\text{Ru}_x\text{O}_4$, the ferromagnetic (FM) ordering in SrRuO_3 (a material closely related to Sr_2RuO_4) and, most importantly, similarities between the normal-state characteristics of

Table 1. The Rice–Sigrist proposal [9] on spin-triplet pairing states in Sr_2RuO_4 with a point group D_{4h} .

| Pairing state | J, J_z | $\mathbf{d}(\mathbf{k})$ | Analog in ^3He |
|----------------------|------------|--------------------------|-------------------------|
| $A_{1u}(\Gamma_1^-)$ | 0, 0 | $xk_x + yk_y$ | B-phase |
| $A_{2u}(\Gamma_2^-)$ | 1, 0 | $xk_y - yk_x$ | B-phase |
| $B_{1u}(\Gamma_3^-)$ | 2, ± 2 | $xk_x - yk_y$ | B-phase |
| $B_{2u}(\Gamma_4^-)$ | 2, ± 2 | $xk_y + yk_x$ | B-phase |
| $E_u(\Gamma_5^-)$ | 1, ± 1 | $z(k_y \pm ik_x)$ | A-phase |

^3He and those of Sr_2RuO_4 , such as the Wilson ratio, strongly favor a p-wave pairing. A similar prediction [10] was made independently on the grounds that $\text{Sr}_2\text{Ir}_{1-x}\text{Ru}_x\text{O}_4$ features an $S = 1$ correlation and Hund’s rule coupling may be at work in Sr_2RuO_4 . Assuming that superconductivity in Sr_2RuO_4 is two-dimensional (2D) in nature, the order parameter should be a function of only the x and y components of the wave vector \mathbf{k} , k_x and k_y , but not the z component, k_z . In this case, five spin-triplet states [9] are allowed. The corresponding forms of the d-vector are listed in table 1. It can be seen that, in the Rice–Sigrist scheme, Γ_{1-4}^- belong to one-component representations with the direction of the d-vector in the ab -plane. The in-plane direction of the d-vector varies on the Fermi surface, corresponding to the B phase [2] of the superfluid ^3He . The Γ_5^- state with a two-component representation, on the other hand, has the direction of the d-vector along the c -axis, which corresponds to the A phase [2] of the superfluid ^3He . Interestingly, the spins of all Cooper pairs in the A phase are in the ab plane—spins are partially ordered.

Many experiments have been carried out on Sr_2RuO_4 to address its pairing symmetry. The Josephson effect and phase-sensitive measurements provided particularly strong support to the picture that Sr_2RuO_4 is a chiral p-wave superconductor. In this paper, I will present a review of the Josephson effect and phase-sensitive measurements carried out so far, focusing on work carried out primarily at Penn State. I will also summarize the physical properties of Sr_2RuO_4 and discuss briefly the eutectic phase of $\text{Ru-Sr}_2\text{RuO}_4$ and several unresolved issues regarding Sr_2RuO_4 .

2. Physical properties of Sr_2RuO_4

Originally synthesized in 1959 [11], Sr_2RuO_4 was rediscovered as a substrate material for the growth of single crystalline films of high- T_c superconductors [12] and as a possible 4d transition metal oxide counterpart of the 3d high- T_c cuprates in the search for novel superconductors [13]. Superconductivity in Sr_2RuO_4 was discovered in 1994, 35 years after the initial synthesis. This discovery generated intense interest in the superconducting materials community because Sr_2RuO_4 is isostructural with the first high- T_c cuprate, $(\text{La, Ba})_2\text{CuO}_4$, and the only transition metal oxide with a layered perovskite crystal structure that becomes superconducting without the presence of Cu. (So far, Sr_2RuO_4 is the only known superconducting ruthenium oxide.) Therefore, it was hoped that the study of Sr_2RuO_4 could provide fresh insight into the mechanism of superconductivity in high- T_c cuprates.

It became clear that Sr_2RuO_4 is very different from high- T_c cuprates. In the normal state, Sr_2RuO_4 is a paramagnetic metal, showing the familiar Fermi liquid behavior rather than the

exotic non-Fermi liquid behavior well known in high- T_c cuprates. Structurally, Sr_2RuO_4 is a quasi-2D material featuring a periodic stacking of a perovskite RuO_2 layer separated by two rock-salt SrO layers. Electronically, Sr_2RuO_4 is one of the most anisotropic metals known, with a ratio of out-of- to in-plane resistivities >200 at room temperature and >800 right above T_c . The Fermi surface of Sr_2RuO_4 consists of three nearly cylindrical sheets [14, 15], including the γ band originating from the d_{xy} , and the α and the β bands from the d_{xz} and d_{yz} orbitals. The α band is hole-like, while the β and γ bands are electron-like. Nuclear magnetic resonance (NMR) measurements suggest that magnetic fluctuation in Sr_2RuO_4 is orbital dependent [16], which is also apparent from magneto-thermoelectrical measurements [17]. Strongly orbital-dependent normal state properties should lead to orbital-dependent superconductivity, as suggested theoretically [18, 19].

Sr_2RuO_4 is the $n = 1$ member of the $\text{Sr}_{n+1}\text{Ru}_n\text{O}_{3n+1}$ Ruddelsden–Popper (R–P) homologous series (figure 1). The $n = \infty$ member of the series, SrRuO_3 , is a 3D ferromagnet with a $T_c^{\text{FM}} = 160$ K [20]. The $n = 5, 4$ and 3 members, $\text{Sr}_6\text{Ru}_5\text{O}_{16}$, $\text{Sr}_5\text{Ru}_4\text{O}_{13}$ and $\text{Sr}_4\text{Ru}_3\text{O}_{10}$, are all layered ferromagnets [21]. $\text{Sr}_4\text{Ru}_3\text{O}_{10}$, the most 2D ferromagnet ($T_c^{\text{FM}} \approx 100$ K) in this R–P series, was found to exhibit some unusual magnetoelastic [22] and magneto-thermoelectric [23] properties. The $n = 2$ member, $\text{Sr}_3\text{Ru}_2\text{O}_7$, is a paramagnetic metal, showing a low-temperature metamagnetic transition at a field between 5 and 8 T, depending on the field orientation [24]. Bulk measurements showed that both FM and antiferromagnetic (AFM) fluctuations are present in $\text{Sr}_3\text{Ru}_2\text{O}_7$ [25, 26], which may be a reflection of incommensurate magnetic fluctuation (IMF) peaked around $(\pm 1/2, 0)(\pi/a)$ and $(0, \pm 1/2)(\pi/a)$, as revealed in the inelastic neutron scattering (INS) measurements [27]. The end compound in the R–P series, Sr_2RuO_4 , features broadly enhanced magnetic fluctuation. INS measurements revealed the presence of IMF with peaks in the susceptibility found around $(\pm 2/3, \pm 2/3)(\pi/a)$ [28]. The IMF appears to originate from the 1D d_{xz} and d_{yz} bands, based on local density approximation calculations [28]. The evolution of the magnetic property within this R–P series appears to suggest that the tendency towards an FM ordering has to be fully suppressed in order for the p-wave superconductivity to emerge, which appears to differ from the commonly held belief that FM fluctuation would help spin-triplet pairing.

Experimental evidence available for unconventional, non-s-wave superconductivity in Sr_2RuO_4 is abundant. Early NMR and nuclear quadruple resonance (NQR) $1/T_1$ studies of Sr_2RuO_4 yielded no Hebel–Slichter coherent peak [29], offering evidence for non-s-wave superconductivity in this material. Measurements on Pb– Sr_2RuO_4 –Pb junctions showed an unexpected drop in the temperature dependence of the critical current [30], $I_c(T)$, suggesting that Sr_2RuO_4 is a type of superconductor different from Pb. The occurrence of superconductivity in Sr_2RuO_4 was found to be extremely sensitive to the presence of impurities [31], again suggesting that this material cannot be an s-wave superconductor. (It is well known that only s-wave superconductivity can survive a substantial amount of disorder. For non-s-wave superconducting pairing, the elastic mean-free path has to be larger than the zero-temperature superconducting coherence length, which is possible typically only when the T_c is high, as in the case of high- T_c superconductors.) Evidence for unconventional, non-s-wave superconductivity was also found in an elastic neutron scattering study that revealed a square rather than a triangular vortex lattice [32], and in tunneling measurements showing the existence of Andreev surface bound states [33, 34].

The presence of nodes in the superconducting order parameter is another hallmark of the unconventional superconductivity. Measurements of the thermodynamic, magnetic and

transport properties in clean, single crystalline Sr_2RuO_4 at temperatures much lower than its T_c showed power-law behavior [35]–[39], suggesting the presence of a large residual density of states (DOS) in the zero-temperature limit. These results would have been a firm indication of the presence of nodes in the superconducting order parameter if Sr_2RuO_4 were a single-band superconductor. However, the presence of multiple bands across the Fermi surface makes it possible that the band-dependent gap is responsible for the large DOS found well below T_c . On the other hand, specific heat measurements with the orientation and magnitude of the magnetic field varied were used [40] to evaluate the node structure in the superconducting parameter of Sr_2RuO_4 , leading to the suggestion that the superconducting order parameter in Sr_2RuO_4 is band dependent with vertical line nodes [40].

Experiments also suggest that Sr_2RuO_4 features a time-reversal symmetry breaking superconducting state, which can be either chiral p- or d-wave. The earliest experimental evidence for such a superconducting state in Sr_2RuO_4 came from the observation of a spontaneous magnetic field in muon spin rotation measurements [41] (a result confirmed by other groups [42, 43]), a large nonzero Kerr rotation below T_c in high-resolution polar Kerr effect measurements [44] and a non-symmetric quantum interference pattern in in-plane Josephson junctions of $\text{Pb-Sr}_2\text{RuO}_4$ [45]. Within the Rice–Sigrist scenario, the only pairing state with such a property is that of Γ_5^- , shown in table 1.

The spin configuration of the superconducting state in Sr_2RuO_4 was first probed by the NMR Knight shift [46] measurements with the magnetic field applied along an in-plane direction, showing that the spin susceptibility is a constant across the T_c . Polarized-neutron scattering measurements [47] led to the same conclusion. NMR measurements on Sr_2RuO_4 with the field aligned along the c -axis are difficult because its c -axis upper critical field is very small. However, recently NMR Knight shift measurements [48, 49] were carried out on Sr_2RuO_4 with a c -axis field as small as 200 G (far below the c -axis critical field). Interestingly, the measurements did not reveal the expected drop in the spin susceptibility below T_c . The result was interpreted in a d-vector rotation scenario—the d-vector is along the c -axis in zero field but rotated to an in-plane direction in a field as small as 200 G—that preserves the spin-triplet pairing picture. This interpretation requires a small spin–orbital coupling. On the other hand, first-principle studies [50] appear to suggest a strong, rather than weak, spin–orbital coupling in Sr_2RuO_4 . Therefore, the implication of the NMR Knight shift results from Sr_2RuO_4 needs to be explored further.

Superconducting quantum interference device (SQUID)-based phase-sensitive measurements [51] probe the variation in the phase of the superconducting order parameter in real or reciprocal space. These measurements on Sr_2RuO_4 showed that the phase of the superconducting order parameter changes by π under a 180° rotation, demonstrating explicitly a p-wave pairing in this superconductor. Combining with the observation of a selection rule of Josephson coupling between Sr_2RuO_4 and an s-wave superconductor [52], the pairing in Sr_2RuO_4 must be that of Γ_5^- , listed in table 1.

3. Josephson coupling between an s- and a p-wave superconductor

Josephson coupling between two superconductors through a tunnel barrier is linked directly to the overlapping integral of the superconducting order parameters of the two superconductors. Therefore, Josephson coupling between an s- and a p-wave superconductor is possible only because of the spin–orbital coupling [53]–[55]. In the absence of the spin–orbital coupling, spin

is a good quantum number; spin-singlet and spin-triplet wave functions are orthogonal with one another with zero overlapping of the wave functions. The Josephson coupling between the s- and the p-wave superconductor would be strictly zero without spin-orbital coupling. In the case of a superconducting weak link, however, Josephson coupling between an s- and a p-wave superconductor is still possible, even without the spin-orbital coupling [56].

The Josephson current density between an s- and a p-wave superconductor through a planar tunnel junction with translational invariance along the junction plane is predicted to be

$$J_s \sim \langle \Psi_s \mathbf{d}(\mathbf{k}) \cdot (\mathbf{k} \times \mathbf{n}) \rangle_{\text{FS}}, \quad (1)$$

where Ψ_s and $\mathbf{d}(\mathbf{k})$ are order parameters for s- and p-wave superconductors, respectively, \mathbf{k} is the wave vector, \mathbf{n} is the interface normal vector and $\langle \dots \rangle_{\text{FS}}$ denotes an appropriate average over the Fermi surface. According to equation (1), Josephson coupling between an s- and a p-wave superconductor through a planar tunnel junction is highly orientation dependent. In particular, if the tunnel junction plane is perpendicular to the direction of the \mathbf{d} -vector, which will make \mathbf{n} parallel to $\mathbf{d}(\mathbf{k})$, J_s is strictly zero, even though the specific k -dependence of the \mathbf{d} -vector is not known. This general conclusion provides a convenient way to check whether Sr_2RuO_4 is indeed consistent with possessing a p-wave pairing, as predicted by theory.

Similar to s-wave superconductors, the strength of the Josephson coupling between an s- and a p-wave superconductor can be measured by the value of $I_c R_N$, where I_c is the critical current and R_N is the normal-state junction resistance. The Josephson coupling between two dissimilar s-wave superconductors at $T = 0$ is given by the Ambegaokar–Baratoff (A–B) formula [57],

$$I_c = (\Delta_1 / R_N e) K([1 - (\Delta_1 / \Delta_2)^2]^{1/2}), \quad (2)$$

where Δ_1 and Δ_2 are the superconducting energy gaps, and the function K is the elliptic integral of the first kind. This result suggests that the critical current of an s-wave Josephson junction is determined only by junction resistance R_N and the superconducting energy gaps of the two superconductors, independent of the details of the junction. In the case where the two superconductors have the same gap, Δ , we have

$$I_c = \pi \Delta / 2e R_N. \quad (3)$$

To calculate the value of $I_c R_N$ for a Josephson junction between an s- and a p-wave superconductor, one needs to know the precise functional forms of $\mathbf{d}(\mathbf{k})$ and the tunneling matrix entering equation (1). In the A–B calculation for s-wave Josephson junctions, the s-wave order parameter is assumed to be independent of \mathbf{k} . (However, even for s-wave superconductors, the superconducting order parameter can, in principle, have anisotropy in \mathbf{k} -space with its sign unchanged on the Fermi surface.) The integration of the tunneling matrices then results in R_N , making the A–B value of $I_c R_N$ depend only on the values of the energy gaps of the two superconductors involved. For a Josephson junction involving a non-s-wave superconductor, similar convenience is not available, making analytic results for J_s difficult to obtain. Numerical calculations [56, 58, 59] for J_s between an s- and a p-wave superconductor have generally yielded values that are much lower than the corresponding A–B value of $I_c R_N$, assuming that the p-wave superconductor is an s-wave with an energy gap that is the same as the maximum gap of the p-wave superconductor.

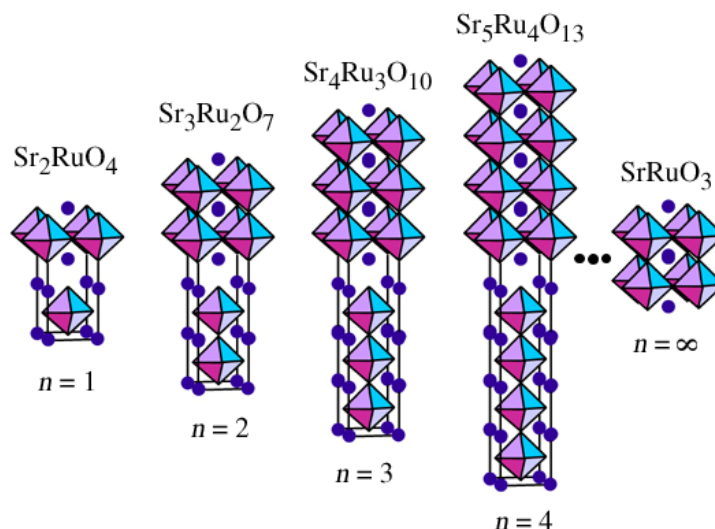


Figure 1. Crystal structure of Sr_2RuO_4 and other compounds in the Ruddlesden–Popper (R–P) homologous series of $\text{Sr}_{n+1}\text{Ru}_n\text{O}_{3n+1}$. Here, n denotes the number of repeating RuO_2 layers in a unit cell. RuO_6 octahedral and Sr atoms (filled circles) are shown schematically.

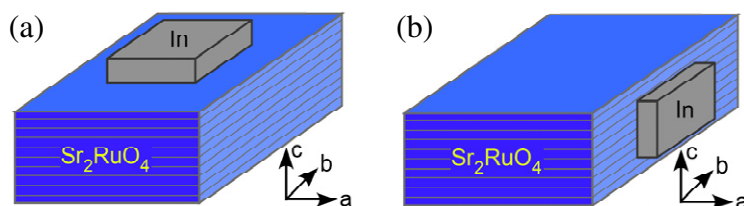


Figure 2. Schematics of samples used in the Josephson coupling selection-rule experiment using In/ Sr_2RuO_4 junctions. The Josephson coupling is zero along the c -axis in (a) and finite (nonzero) for in-plane directions in (b).

4. Experimental studies of the Josephson effects of Sr_2RuO_4

4.1. Selection rule

Experimentally, the Josephson coupling between an s-wave superconductor In and Sr_2RuO_4 was measured in c -axis and in-plane junctions prepared by pressing freshly cut pure In wire directly onto a cleaved ab or polished ac face of Sr_2RuO_4 [52], as shown schematically in figure 2. The Josephson coupling for the in-plane In/ Sr_2RuO_4 junctions was found to be finite (figure 3(a)). The temperature dependence of the critical current in this Josephson junction, an example of which is shown in figure 3(b), was found to vary from sample to sample, probably as a result of an inhomogeneous junction. Other consequences of the junction inhomogeneity will be discussed in section 4.3.

None of the pressed In junctions prepared on the cleaved ab face was found to show a finite supercurrent. The absence of a finite supercurrent does not seem to be due to a suppressed $I_c R_N$ because the value of $I_c R_N$ for in-plane tunnel junctions was

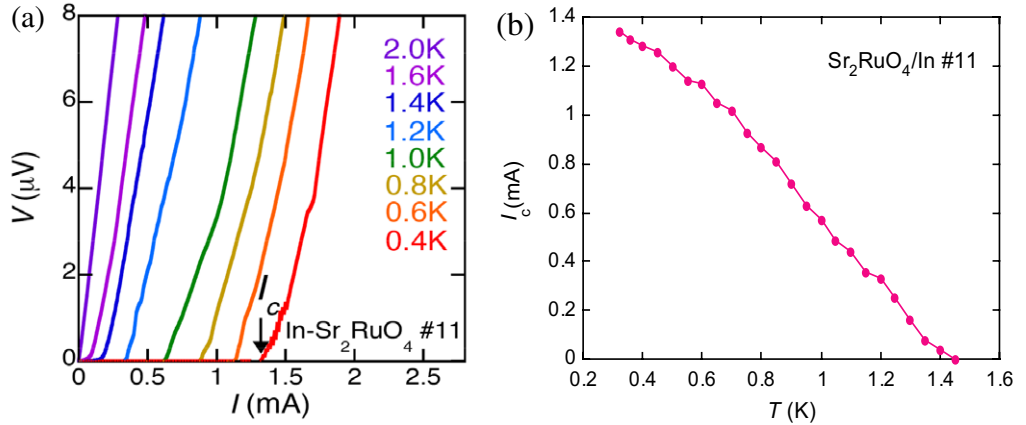


Figure 3. (a) Current (I)–voltage (V) curves of an in-plane In/Sr₂RuO₄ junction at various temperatures (T), as indicated. The critical current, I_c , is defined by the onset of a finite voltage. (b) I_c versus T for an in-plane junction with $R_N = 0.12 \, \Omega$.

found to be large (see below). It is known that superconductivity is suppressed on the ab face of Sr₂RuO₄ because of the rotation of RuO₆ octhedra [59]. However, the number of RuO₂ layers that are subject to the suppression of superconductivity should not be more than a few unit cells, based on elastic energy considerations. The s-wave superconductor (S) In, the normal (N) region near the ab surface and superconducting bulk Sr₂RuO₄ single crystal (S') should form an SNS' planar Josephson junction by the proximity effect. A finite Josephson coupling between the two superconductors is expected, as long as the N-region is within a few times the normal coherence length in the clean limit, ξ_N . This length, $\xi_N = \hbar v_F^c / 2\pi k_B T$, where v_F^c is the Fermi velocity along the c -axis ($v_F^c = 1.4 \times 10^4 \text{ m s}^{-1}$ [15]), is $\xi_N \approx 80 \text{ nm}$ for Sr₂RuO₄ at $T = 0.3 \text{ K}$, the lowest temperature for this set of measurements. This number is larger than the c -axis lattice constant $c = 1.28 \text{ nm}$ by almost two orders of magnitude. It is unlikely that the N-layer formed on a freshly cleaved Sr₂RuO₄ single crystal can be so thick that the supercurrent in c -axis In/Sr₂RuO₄ junctions vanishes. On the other hand, the above result of a selection rule of the Josephson coupling between In and Sr₂RuO₄ is consistent with the \mathbf{d} -vector of Sr₂RuO₄ aligned along the c -axis, which is the Γ_5^- state within the Rice–Sigrist scheme (table 1), according to equation (1).

4.2. Strength of the Josephson coupling

The strength of the Josephson coupling can be measured by the $I_c R_N$ value, as pointed out above. However, even for two s-wave superconductors, experimentally the A–B limit given in equations (2) or (3) often represents only the upper limit of the Josephson coupling if the bulk gap values are used. The typical interpretation of this observation is that the superconducting energy gaps may be suppressed on the surface, causing I_c to fall below the bulk values. For in-plane In/Sr₂RuO₄ junctions, no acceptable value for the energy gap of Sr₂RuO₄ is available. However, if one estimates the gap values from T_c using the BCS result, $\Delta = 1.76 k_B T_c$, an A–B limit of 0.516 mV would be obtained for an In/Sr₂RuO₄ junction in the zero-temperature limit, assuming that Sr₂RuO₄ is an s-wave superconductor. At $T = 0.3 \text{ K}$, the values of $I_c R_N$ were

Table 2. $I_c R_N$ values for several In–Sr₂RuO₄ junctions obtained at the measurement temperature, T_{meas} . Estimated A–B limit, $I_c R_N^{\text{A-B}}$, is also shown for comparison.

| Sample | T_{meas} (K) | I_c (mA) | R_N (Ω) | $I_c R_N^{\text{A-B}}$ (mV) | $\frac{I_c R_N}{I_c R_N^{\text{A-B}}}$ |
|--------|--------------------------|---------------|-----------------------|--------------------------------|--|
| #12 | 0.34 | 1.8 | 0.10 | 0.52 | 0.35 |
| #11 | 0.34 | 1.3 | 0.12 | 0.52 | 0.20 |
| #9 | 0.32 | 0.58 | 0.18 | 0.52 | 0.20 |
| #5 | 0.40 | 0.50 | 0.09 | 0.52 | 0.09 |

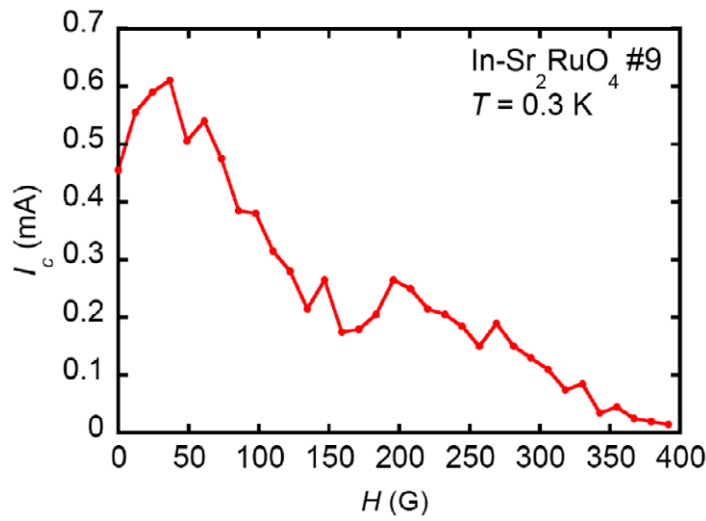


Figure 4. The magnetic field dependence of the critical current (I_c) for an In/Sr₂RuO₄ junction, plotted as a function of magnetic field (H).

found to be 0.10 mV for an In/Sr₂RuO₄ sample, shown in figure 3(b), a substantial fraction of the A–B limit. Here, the value was taken as the junction resistance measured at the T_c of In because the normal-state junction resistance is slightly temperature dependent. Similar results were observed in other samples (table 2). Because the sign changes in $d(\mathbf{k})$ tend to reduce J_s while $\langle \dots \rangle_{\text{FS}}$ is carried out in equation (1), as pointed above [56, 58, 59], a substantial fraction of the A–B limit for Josephson coupling between an s- and a p-wave superconductor is not expected. This issue is yet to be resolved.

4.3. Magnetic field dependence

For a Josephson junction, I_c will oscillate as a function of magnetic field applied along the junction plane with a period of $H_0 = \Phi_0/A$, where A is the junction area given by $W(\lambda_1 + \lambda_2)$, W is the dimension of the junction, and λ_1 and λ_2 are the penetration depths of the two superconductors. A Fraunhofer diffraction pattern is also expected in $I_c(H)$, and its amplitude drops quickly after the first few periods. For an In–plane In–Sr₂RuO₄ junction, $\lambda_1 = 64$ nm and $\lambda_2 = \lambda_{\text{ab}} = 180$ nm. If the size of the In–Sr₂RuO₄ junction is similar to that of the In wire, ~ 1 mm, then H_0 will be a fraction of a Gauss. In figure 4, the value of I_c for an In/Sr₂RuO₄

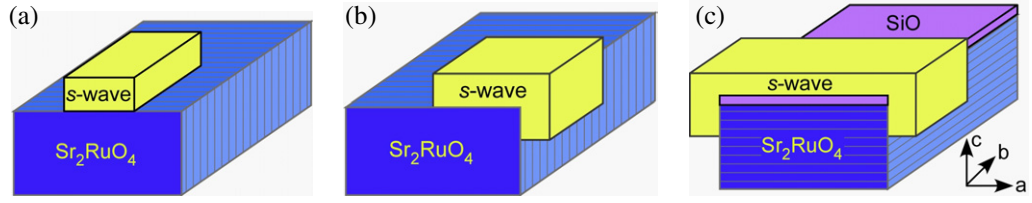


Figure 5. Schematics of phase-sensitive devices for determining the pairing symmetry of Sr_2RuO_4 : (a) same-side junction, (b) corner junction and (c) opposite-side SQUID. A magnetic field is applied along the junction plane.

Table 3. Expected features in the quantum interference pattern of the critical current I_c as a function of the total flux, Φ , for devices shown in figure 5.

| Pairing state | Same-side junction | Corner junction | Opposite SQUID |
|----------------|------------------------------|----------------------------------|------------------------------|
| <i>s</i> -wave | $I_c(\Phi = 0) = \text{max}$ | $I_c(\Phi = 0) = \text{max}$ | $I_c(\Phi = 0) = \text{max}$ |
| <i>d</i> -wave | $I_c(\Phi = 0) = \text{max}$ | $I_c(\Phi = 0) = \text{min}$ | $I_c(\Phi = 0) = \text{max}$ |
| <i>p</i> -wave | $I_c(\Phi = 0) = \text{max}$ | $I_c(\Phi = 0) = \text{neither}$ | $I_c(\Phi = 0) = \text{min}$ |

junction is plotted as a function of H . However, neither a Fraunhofer pattern nor a regular field modulation in $I_c(H)$ was observed. The above observation is consistent with the behavior of a non-uniform Josephson junction with a size of the order of microns. This result is not surprising, however, given that a mechanically polished ac face of Sr_2RuO_4 possesses unavoidable mechanical damage and therefore disorder. Nevertheless, equation (1) is valid [55] so long as the translational invariance can be maintained over the superconducting coherence lengths in the zero-temperature limit, $\xi(0)$, which are 66 and 3.3 nm for Sr_2RuO_4 (along the in- and out-of-plane directions, respectively) and 44 nm for bulk In. Therefore, the selection rule result discussed above appears to be unaffected.

An interesting observation is that the Josephson current does not vanish until 400 G, larger than the minimal field required for the \mathbf{d} -vector to rotate in Sr_2RuO_4 , 200 G [48, 49]. If the \mathbf{d} -vector does rotate to the in-plane direction at a field as small as 200 G, suggested by the NMR Knight shift measurements, one would expect the Josephson coupling to vanish or undergo a change at a characteristic magnetic field near or below 200 G. While the data shown in figure 4 appear to show a feature between 150 and 200 G, more data from a systematic study are needed to draw a conclusion.

5. Phase-sensitive experiments on bulk Sr_2RuO_4

In a phase-sensitive experiment, the phase rather than the amplitude of the superconducting order parameter is determined as a function of crystal orientation. Geshkenbein, Larkin and Barone (GLB) proposed the original phase-sensitive-measurement idea, including both the SQUID and the tricrystal configurations, in the context of detecting *p*-wave superconductivity in heavy Fermion superconductors [60]. Leggett rediscovered the SQUID-based phase-sensitive measurements for *d*-wave superconductors in the high- T_c research [61]. For high- T_c superconductors, the symmetry of the order parameter was determined unambiguously

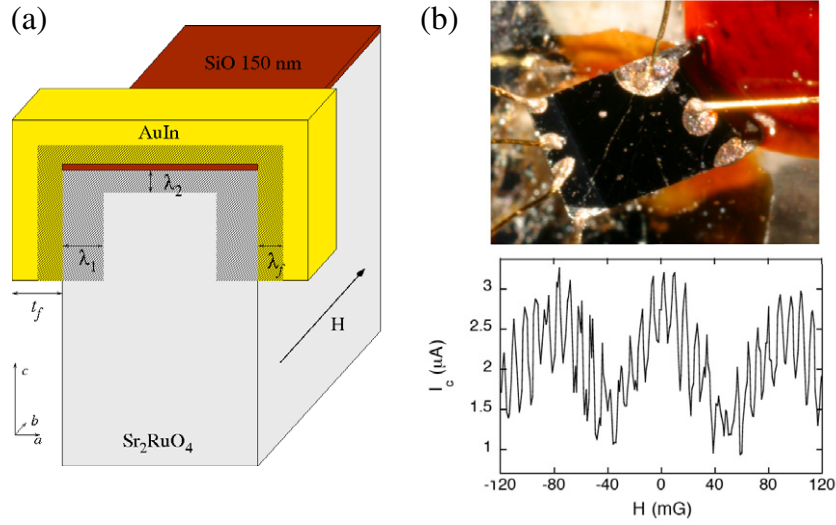


Figure 6. (a) Schematic of a GLB $\text{Au}_{0.5}\text{In}_{0.5}\text{--Sr}_2\text{RuO}_4$ SQUID. (b) Upper panel: optical picture of an $\text{Au}_{0.5}\text{In}_{0.5}\text{--Sr}_2\text{RuO}_4$ SQUID. The two junctions are on the lower and upper left sides of the crystal. (Photo credit: John Passaneau.) Lower panel: example of quantum oscillation in $I_c(H)$ of a GLB SQUID shown in (a).

only after phase-sensitive experiments were carried out [62, 63]. Similarly, phase-sensitive measurements of Sr_2RuO_4 are needed in order to settle the pairing symmetry of Sr_2RuO_4 experimentally.

Our approach to phase-sensitive measurements of Sr_2RuO_4 is to build a phase-sensitive toolkit, as illustrated in figure 5. According to equation (1), the Josephson currents flowing through the two Josephson junctions prepared on the opposite faces of a spin-triplet superconductor (the two junctions have normal vectors in \mathbf{n} and $-\mathbf{n}$, respectively) of the GLB SQUID (figure 5(c)) are out of phase with one another by 180° , the intrinsic phase difference of the superconducting order parameter after a rotation by π . Similarly, for a corner or a same-face SQUID, the intrinsic phase difference will be 90° or 0° , respectively. Experimentally, however, a single junction on a side or a corner of the crystal will work for the same purpose. Single Josephson junctions have smaller effective area for modulating magnetic flux than the SQUID, making the device less susceptible to flux trapping (see below). The expected experimental signatures for the three possible pairing symmetries in the quantum interference pattern are shown in table 3. It should be emphasized that the flux threaded in the SQUID loop (or the Josephson junction) is the total amount of flux, different in general from the applied flux (see below).

To prepare the phase-sensitive experiment toolkit for Sr_2RuO_4 , we used single-crystal based structures (figure 6) since superconducting epitaxial films of Sr_2RuO_4 were not (and are still not) available [64]. The s-wave superconductor, $\text{Au}_{0.5}\text{In}_{0.5}$, with a T_c of $\sim 0.5 \text{ K}$ was used because it wets Sr_2RuO_4 crystal well. In addition, it possesses a long superconducting coherence length ($\xi(0) > 150 \text{ nm}$) that favors the establishment of a Josephson coupling with Sr_2RuO_4 . All of our $\text{Au}_{0.5}\text{In}_{0.5}\text{--Sr}_2\text{RuO}_4$ junctions (SQUIDS) feature a naturally formed tunnel barrier.

Several important experimental issues were encountered in carrying out the phase-sensitive measurements [51]. Firstly, the preparation of the Josephson junction or SQUID structures

requires mechanical polishing of the crystal in order to have the desired Josephson junction planes. Because of the extreme sensitivity of superconductivity to disorder in Sr_2RuO_4 , mechanical polishing clearly has a negative effect on superconductivity, consistent with the observation that only a small fraction of our samples were found to display a measurable supercurrent. Secondly, Ru islands associated with the eutectic phase of Ru– Sr_2RuO_4 with an onset superconducting transition temperature of nearly 3 K (see below) were commonly found in a polished crystal surface. These Ru islands are not desirable to avoid unnecessary complications. To do so, every polished surface was carefully inspected under an optical microscope. Thirdly, the applied flux, Φ_{ext} , used to modulate the critical current, I_c , needs to be close to the total flux (Φ) threading the SQUID. Finally, additional complications can result from the formation of domains of $k_x + ik_y$ and $k_x - ik_y$, associated with a chiral pairing state, such as the Γ_5^- state, listed in table 1. The last two issues will be discussed in more detail below.

To ensure that the total flux threading the SQUID or the junction (Φ) is as close as possible to the applied flux (Φ_{ext}), it is useful to note that the total flux is given by $\Phi = \Phi_{\text{ext}} + \Phi_{\text{ind}} + \Phi_{\text{trap}} + \Phi_{\text{bkgd}}$, where Φ_{ind} is the induced flux, Φ_{trap} the trapped flux and Φ_{bkgd} the background flux. Clearly, Φ_{ind} , Φ_{trap} , and Φ_{bkgd} all need to be minimized. Among them, Φ_{bkgd} is the easiest to deal with—it can be minimized by careful magnetic shielding. Φ_{ind} is determined by the sample size and the asymmetry of the SQUID. For an opposite-face SQUID sample, $\Phi_{\text{ind}} = LI_{\text{circ}} = L(I_1 - I_2)$, where I_{circ} is the circulating current in the loop and L is the self-inductance [65]. Early SQUID-based phase-sensitive experiments [66, 67] on high- T_c superconductors relied on an extrapolation of $R(H)$ measured at currents above I_c to obtain the values at zero current, an approach criticized by others [68] and apparently abandoned in favor of the corner junction experiments [69, 70]. We adopted an alternative approach by showing that $I_c(\Phi_{\text{ext}} = 0)$ corresponds to a minimum close to T_c of the SQUID. In this case, $I_{\text{circ}} \rightarrow 0$, so that $\Phi = \Phi_{\text{ext}} + \Phi_{\text{ind}} \rightarrow \Phi_{\text{ext}}$, if $\Phi_{\text{trap}} = 0$. It should be noted that Φ_{trap} , which could make conventional SQUIDS mimic the behavior of an unconventional SQUID [71]–[73], can take up an arbitrary value. In general, the fluxoid quantization requires that $2\pi m = \phi_1 - \phi_2 + (2\pi/\Phi_0)(\Phi_{\text{ext}} + \Phi_{\text{ind}} + \Phi_{\text{trap}})$, where m is an integer (or 0), and ϕ_1 and ϕ_2 are phase drops across the two junctions in the SQUID. Clearly, ϕ_1 and ϕ_2 , the two degrees of freedom of the system, can adjust themselves to accommodate any arbitrary Φ_{trap} . Trapped flux leads to an asymmetric envelope of the $I_c(H)$, which was used to determine whether flux is trapped in our SQUIDS. We found that warming up and cooling down the sample in zero field slowly appeared to help prepare a trapped-flux-free state in our SQUIDS.

In the case that Sr_2RuO_4 features the Γ_5^- state listed in table 1, it is important that a procedure is found so that our samples involve only a single or known number of domains. However, domains have not been observed directly in Sr_2RuO_4 . Therefore, a safe strategy would be to work to prepare a single-domain state assuming that domains do exist. A possible way for the domains to form is through a slight variation in either the superconducting transition temperature or the sample temperature that leads to nucleation of superconducting regions in isolated spots as the temperature is lowered. To minimize this tendency, we cooled the sample at a very slow (\sim hours), computer-controlled rate in our experiment, which should help us to minimize the chances of having multiple domains as well as the trapped flux. Obviously, further work is needed to detect and control the formation of domains.

Data taken close to T_c of the GLB SQUIDS (figure 7) demonstrated that the phase of the order parameter changes by π after 180° rotation, providing compelling evidence that Sr_2RuO_4 is indeed an odd-parity, spin-triplet superconductor [51]. These results, together with

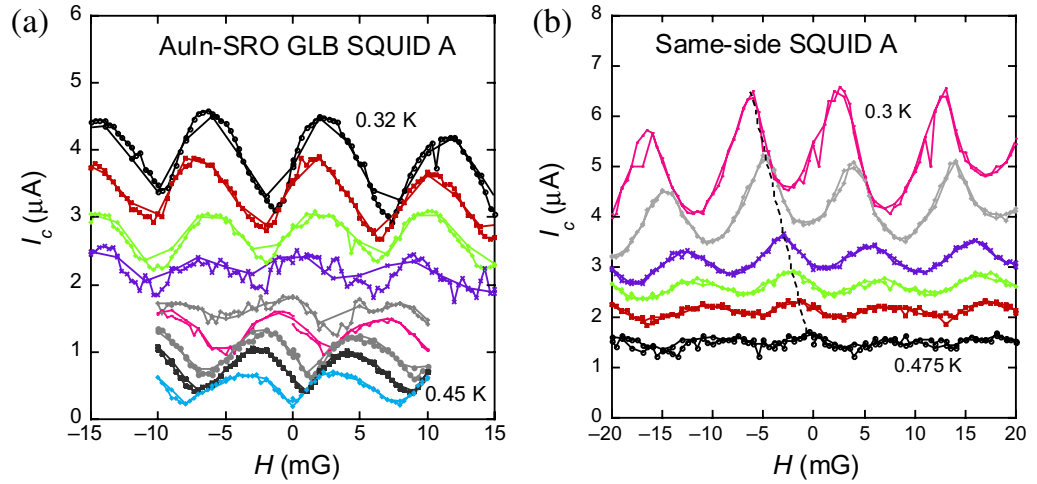


Figure 7. $I_c(H)$ for an opposite side (a) and the same side (b) of AuIn– Sr_2RuO_4 SQUID prepared on Sr_2RuO_4 single crystals.

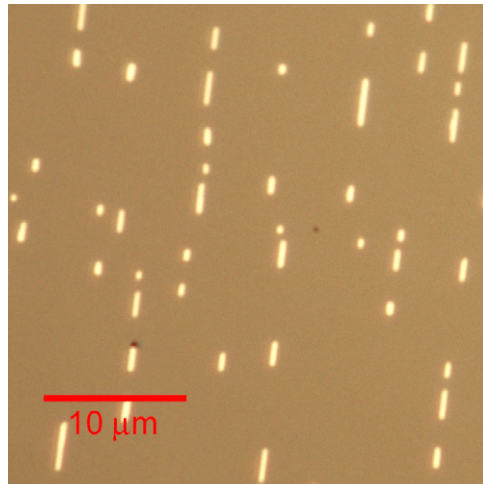


Figure 8. Optical image of a mechanically polished crystal surface of Ru– Sr_2RuO_4 single crystal. The bright regions are single-crystal islands of Ru.

the previous Josephson selection rule results discussed above, showed that the pairing symmetry in Sr_2RuO_4 is that of Γ_5^- state within the Rice–Sigrist scheme listed in table 1.

6. Superconductivity in the eutectic phase of Ru– Sr_2RuO_4

Superconducting single crystals of Sr_2RuO_4 are synthesized by the floating zone method. Because of the high volatility of Ru, it is necessary to compensate for the Ru loss during the growth by adding extra RuO_2 in the starting rod. A eutectic phase featuring single-crystalline islands of pure Ru metal embedded in the single-crystal matrix of Sr_2RuO_4 (figure 8) is frequently found to form in the crystal, especially close to the center of the crystal rod [74]. Surprisingly, superconductivity near the Ru– Sr_2RuO_4 eutectic phase was found to feature

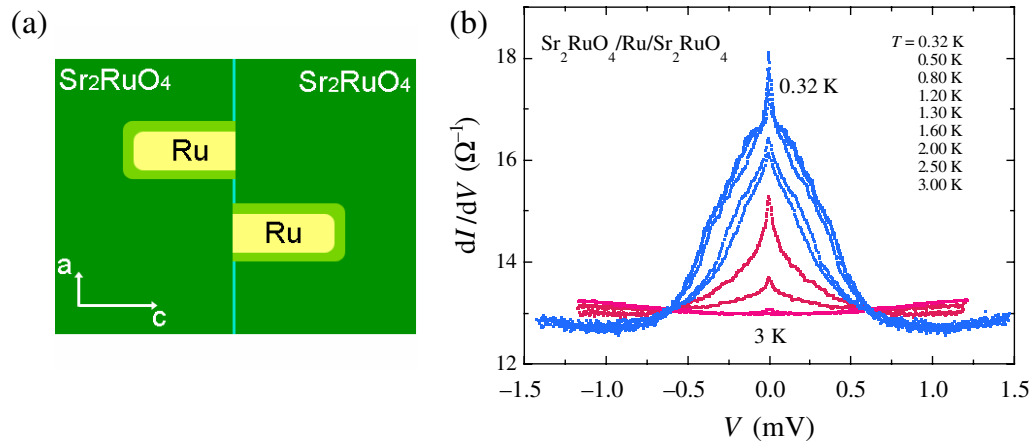


Figure 9. (a) Schematic of a break junction. The 3-K phase is on the Sr_2RuO_4 side of the Ru– Sr_2RuO_4 interface, shown in light green. Away from the Ru islands, the cleaved surface is non-superconducting, resulting in a normal layer (not shown). (b) Tunneling spectra at various temperatures, as indicated.

an unexpected onset T_c as high as 3 K, which was suggested to originate in regions on the Sr_2RuO_4 side of the Ru/ Sr_2RuO_4 interface, based on the anisotropic properties of this so-called 3-K phase. Enhanced superconductivity is known in other interface systems, such as at the atomically sharp interface between Ag and Ge, even though neither Ag nor Ge is superconducting [75]. However, the enhanced superconductivity near the Ru– Sr_2RuO_4 interface was still a surprise, given that the occurrence of superconductivity in Sr_2RuO_4 is sensitive to disorder, including structural imperfections. The interface between two different materials would also appear to function as a pair breaker, even if no disorder is present, which would tend to suppress rather than enhance superconductivity.

In addition, questions about the nature of the 3-K phase, such as whether its pairing symmetry is also p-wave, were raised. To address these issues, we carried out tunneling measurements, which may be the most effective way to address these issues, given that the 3-K phase occurs only near the interface region. On the surface of a non-s-wave superconductor, the intrinsic orientation dependence of the phase of the order parameter results in mid-gap Andreev bound states and an associated zero-bias conductance peak (ZBCP) in the tunneling spectrum [76]–[79], as seen in high- T_c cuprates [80, 81]. Andreev surface bound states were also detected in the bulk phase of Sr_2RuO_4 [33]. However, the fitting to the data may be problematic because the superconducting energy gap obtained from the fitting is unreasonably large.

We prepared break tunnel junctions by cleaving an Sr_2RuO_4 single crystal containing a Ru island (figure 9) [34]. In this sample configuration, the tunneling current will be dominated by conducting channels near the Ru islands, as shown by a ZBCP persisting up to 3 K. The ZBCP marks the presence of ABSs, suggesting that the eutectic phase is an unconventional, non-s-wave superconductor. Theoretically, a p-wave state with horizontal line nodes was found to yield a single peak near the zero bias voltage [82], which can actually fit our data quantitatively. On the other hand, the presence of horizontal nodes appears to have been ruled out by magnetic field-dependent specific heat results [40]. More work is needed to resolve this inconsistency.

The 3-K phase may offer insights into the mechanism of superconductivity in Sr_2RuO_4 . Sigrist and Monien [83] developed a phenomenological theory for the 3-K phase and argued that superconductivity will nucleate in the interface region between Ru islands and the bulk

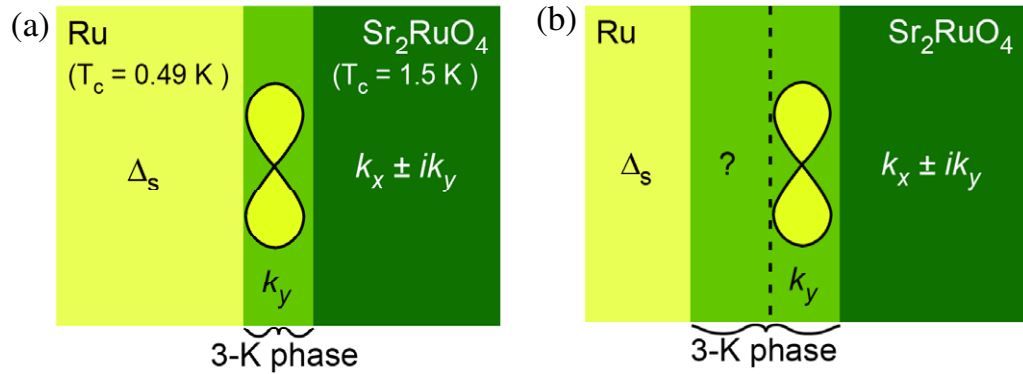


Figure 10. Schematics illustrating the nature of superconductivity in the 3-K phase. The Ru region is shown in yellow and the bulk Sr₂RuO₄ is shown in green, with light green indicating the 3-K phase region. The physical boundary between the Ru island and Sr₂RuO₄ is assumed to be at $x = 0$ (x -axis is along the horizontal direction). The two alternative pictures are illustrated (see text).

Sr₂RuO₄ at a temperature above the bulk T_c (figure 10(a)). It was shown that energetic considerations favor a p-wave with a line node parallel to the normal vector with positive and negative lobes parallel to the interface (say, k_y -state). As the temperature is further lowered, the second component will emerge, forming a time-reversal symmetry breaking state ($k_x \pm ik_y$ -state). However, our recent tunneling measurements [84] did not reveal a proximity induced p-wave superconducting energy gap in the interior of the Ru island, suggesting an alternative picture in which the 3-K phase originates in the region somewhere away from the interface, as shown in figure 10(b).

7. Discussion

Several issues regarding superconductivity in Sr₂RuO₄ remain unresolved. For example, all states listed in table 1 have an isotropic (full) gap. The observed power-law behaviors described above can be attributed to horizontal/vertical nodes in the superconducting order parameter [85]–[89], or orbital-dependent superconductivity (ODS) [18, 19]. In the former case, the vertical nodes would imply that the order parameter dependence is independent of k_z , whereas the horizontal nodes require that the order parameter depends on k_z . In this regard, magnetic field-dependent specific heat measurements seem to rule out k_z dependence of the \mathbf{d} -vector featuring the presence of horizontal nodes [40]. However, the presence of vertical nodes appears to be inconsistent with the tunneling results [34, 82]. Josephson tunneling measurements, which are currently under way, can provide an independent check on the k_z dependence of the \mathbf{d} -vector.

Even though most experiments suggest that Sr₂RuO₄ is a chiral p-wave superconductor represented by the Γ_5^- state, the phase diagram obtained with a precisely aligned in-plane magnetic field [90] does not agree with the theoretical expectations for a chiral p-wave [91]. Furthermore, domains corresponding to $k_x + ik_y$ or $k_x - ik_y$ states and domain walls between them have not yet been observed directly experimentally as pointed out above [92]. Possible sizes of the domains inferred indirectly from various measurements vary greatly [93], adding to the confusion over this issue.

The mechanism of superconductivity of Sr_2RuO_4 is not yet understood. Models based on FM fluctuation [94], AFM fluctuation [95], spin–orbital coupling [96] or Hund’s rule coupling [10] have been proposed. The systematic tests on the proposed mechanisms, which are yet to be carried out, are needed. The eutectic phase of $\text{Ru–Sr}_2\text{RuO}_4$, which may provide insight into the mechanism of superconductivity in Sr_2RuO_4 because of the unexpected enhancement of T_c , needs to be studied further.

8. Conclusion

In this brief review, I have summarized our Josephson tunneling and phase-sensitive measurements of Sr_2RuO_4 . This work represents an important step towards the establishment of an electronic counterpart of superfluid ^3He in Sr_2RuO_4 featuring odd-parity, spin-triplet superconductivity. Further work is needed to determine the precise symmetry form of the superconducting order parameter, and to establish the mechanism of superconductivity in this material.

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